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Sir:

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For: RUBBER COMPOSITION

XX   Specification (26 pages)

XX   _2_ sheets of drawings

XX   Declaration and Power of Attorney

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<u>RUBBER COMPOSITION</u>

BACKGROUND OF THE INVENTION

5 1. Field of the Invention

The present invention relates to a rubber composition, more specifically relates to a rubber composition suitable for use for a tire tread, sidewalls, or other rubber parts and having improved tensile
10 strength, elongation, and abrasion resistance.

2. Description of the Related Art

In recent years, improvements in various types of performance have been sought in the rubber compositions for automobile and other tires etc.
15 Therefore, in tire tread rubber and the like, several types of polymers have been used by blending together. However, when these polymers are incompatible with each other, phase separated interfaces are present. In most cases, these interfaces become starting points for
20 breakage and are believed to have a detrimental effect on the tensile strength, tear strength, abrasion resistance, etc. However, in tire and other rubber products, since the special process of vulcanization is included, it is not possible to apply as is the molecular design of the
25 block copolymer for control of the phase structure as is normally done in rubber/resin blends or resin/resin blends. Therefore, the problem of the phase separation interface of rubber/rubber blends has not been sufficiently studied and no method for solving this
30 problem had been found yet.

In the past, the decrease in the breaking strength due to the incompatibility of polymer blends obtained by blending block copolymers has not been sufficiently studied. Blending, into a blend of natural
35 rubber (NR)/polybutadiene rubber (BR), a small amount of a block copolymer of polybutadiene (BR) and polyisoprene (IR) has only been described slightly in *J. Apply. Polym.*

Sci., 49 (1993) and *RCT.* 66 (1993). However, the compositions of the block copolymers used in these references have insufficient compatibility with BR, and therefore, are not satisfactory in performance for practical use. Further, attempts have been made to add cis-BR into an incompatible polymer blend of cis-BR/SBR so as to improve the abrasion resistance, but the wet braking performance is decreased, and therefore, there is a limit to the amount of addition of cis-BR and there were consequently problems in practical use. In addition, except for the proposals made by the inventors of the present invention (Japanese Unexamined Patent Publication (Kokai) Nos. 7-188510, 8-134267, 8-193147, 8-193146, 8-193145, 8-283465, 8-302071, 10-007844, and 10-036465), examples of blending a block polymer into a rubber composition as a compatibilizing agent have not been known. The previous proposals by the present inventors did not clarify the relationship between the rubber component forming the matrix of the rubber composition and the molecular weight of the block polymer added. Later study resulted in clarification of this point and the present invention has been completed.

SUMMARY OF THE INVENTION

Accordingly, the object of the present invention is to provide a rubber composition capable of eliminating the above-mentioned problems in the prior art and improving the tensile strength, elongation, abrasion resistance, etc. thereof.

In accordance with the present invention, there is provided a rubber composition comprising (i) an incompatible polymer blend comprising at least two diene rubbers selected from the group consisting of rubbers containing at least one conjugated diene monomer and optionally at least one aromatic vinyl monomer, such as natural rubber (NR), polyisoprene rubber (IR), polybutadiene rubber (BR), styrene-butadiene copolymer rubber (SBR), styrene-isoprene copolymer rubber (SIR) and

styrene-isoprene-butadiene rubber (SIBR) and forming two incompatible polymer phases (A) and (B) and (ii) 0.1 to 20 parts by weight, based upon 100 parts by weight of the total polymer component including the block copolymer, of a block copolymer having at least two mutually incompatible blocks (a) and (b), the block (a) being compatible with the polymer phase (A) and being incompatible with the polymer phase (B) and the block (b) being compatible with the polymer phase (B) and incompatible with the polymer phase (A), and comprising at least one conjugated diene monomer (e.g., isoprene, butadiene) and, optionally, at least one aromatic vinyl monomer (e.g., styrene), wherein the molecular weights of the polymers forming the polymer phases (A) and (B) satisfy the following equations (I) and (II):

$$S_A = Mw_{30}(A)/Mw(a) \leq 1.2 \qquad (I)$$

$$S_B = Mw_{30}(B)/Mw(b) \leq 1.2 \qquad (II)$$

wherein $Mw_{30}(A)$: molecular weight of the low molecular weight portion of the polymer forming the polymer phase (A),

$Mw_{30}(B)$: molecular weight of the low molecular weight portion of the polymer forming the polymer phase (B),

$Mw(a)$: weight average molecular weight of block (a) of block copolymer, and

$Mw(b)$: weight average molecular weight of block (b), of block copolymer.

In accordance with the present invention, there is also provided a rubber composition, wherein 5 to 200 parts by weight, based upon 100 parts by weight of the block copolymer, of a polymer (α) compatible with the block (a) and the polymer phase (A) and/or a polymer (β) compatible with the block (b) and polymer phase (B) are further blended and the weight average molecular weights of the polymers (α) and (β) satisfy the following

equations (III) and (IV):

$$S_\alpha = Mw(\alpha)/Mw(a) \leq 1.2 \qquad (III)$$

$$S_\beta = Mw(\beta)/Mw(b) \leq 1.2 \qquad (IV)$$

wherein $Mw(\alpha)$: weight average molecular weight of polymer (α),

$Mw(\beta)$: weight average molecular weight of polymer (β),

$Mw(a)$: weight average molecular weight of block (a) of block copolymer, and

$Mw(b)$: weight average molecular weight of block (b) of block copolymer.

In accordance with the present invention, there is further provided a rubber composition comprised of a block copolymer having at least two mutually incompatible blocks (a) and (b) and comprising at least one conjugated diene and, optionally, at least one aromatic vinyl monomer based upon 100 parts by weight of the same, 5 to 200 parts by weight of a polymer (α) compatible with the block (a) and/or a polymer (β) compatible with the block (b), the weight average molecular weights of the polymers (α) and (β) satisfying the following equations (III) and (IV):

$$S_\alpha = Mw(\alpha)/Mw(a) \leq 1.2 \qquad (III)$$

$$S_\beta = Mw(\beta)/Mw(b) \leq 1.2 \qquad (IV)$$

wherein $Mw(\alpha)$: weight average molecular weight of polymer (α),

$Mw(\beta)$: weight average molecular weight of polymer (β),

$Mw(a)$: weight average molecular weight of block (a) of block copolymer, and

$Mw(b)$: weight average molecular weight of block (b) of block copolymer.

BRIEF DESCRIPTION OF THE DRAWINGS

- 5 -

The present invention will be better understood from the description set forth below with reference to the accompanying drawings, wherein

Fig. 1 is a view of an example of a molecular weight distribution curve (integrated molecular weight curve) of molecular weights measured by GPC forming the basis for finding the molecular weights of the low molecular weight portions of the polymers of the polymer phases (A) and (B) of equations (I) and (II);

GPC Measurement Conditions

GPC: HLC-8020 made by Toso

Column: GMH-HR-H, 2

Temperature: 40°C

Mobile phase: THF

Standard substance: 10 points used between standard polystyrene 1000 to 10,000,000

Approximation method: By tertiary method.

Preparation of polymer sample: 50 mg of the polymer was dissolved in 10 cc of THF. The mixture was stirred at room temperature for about 168 hours so as to dissolve. This was then filtered by a 0.5 micron filter (H25-5 made by Toso) to remove the insolubles. The result was used as the sample. The amount injected into the GPC was made 400 μl.

Fig. 2 is a view of an integrated molecular weight curve obtained by converting the molecular weight distribution curve of Fig. 1, wherein $Mw_{30}(A)$ and $Mw_{30}(B)$ of equations (I) and (II) are found from the molecular weight of the cumulative area 30% as shown in Fig. 2.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

The present inventors found that the above-mentioned object can be achieved by compounding, into a rubber composition formed from at least two rubber phases comprising at least two incompatible rubbers, a block polymer comprising at least two incompatible blocks having molecular weights which are defined by two types of relationships with the molecular weights of the

rubbers forming the two rubber phases.

The tire rubber composition according to the present invention can be obtained by blending (i) an incompatible polymer blend of two polymer phases (A) and (B) comprising at least two types of incompatible rubbers of NR, IR, BR, SBR, SIR, and SIBR, preferably in the weight ratio of (A)/(B) of 90/10 to 10/90, more preferably 85/15 to 15/85) and (ii) 0.1 to 20 parts by weight, preferably 1 to 15 parts by weight, based upon 100 parts by weight of the entire rubber component including a block copolymer, of the block copolymer having at least two blocks comprising monomers selected from isoprene, butadiene, and styrene, wherein the blocks (a) and (b) are mutually incompatible, the block (a) is compatible with the polymer phase (A) and incompatible with the polymer phase (B), and the block (b) is compatible with the polymer phase (B) and incompatible with the polymer phase (A), preferably the weight ratio of (a)/(b) of 80/20 to 20/80, more preferably 60/40 to 40/60, and wherein the molecular weights of the polymers forming the polymer phases (A) and (B) satisfy the equations (I) and (II). Note that S_A and S_B preferably are 0.1 to 1.2, more preferably 0.3 to 1.0.

If the value of S_A and the value of S_B are more than 1.2, the block copolymer added while mixing the rubber component does not disperse well and the effect as a compatibilizing agent cannot be sufficiently exhibited.

Note that the molecular weights of the low molecular weight portions of the polymers forming the polymer phases (A) and (B) mean those found as values of molecular weights (i.e., $Mw_{30}(A)$ and $Mw_{30}(B)$) corresponding to 30% of the cumulative area when converting the curve of the distribution of the molecular weight measured by GPC such as shown in Fig. 1 to the integrated molecular weight curve, as shown in Fig. 2. The GPC is measured by, for example, dissolving the polymer sample into THF, removing the insoluble gel

component by a 0.5 micron filter, then calculating the molecular weight by an equation obtained from the amount of elution of standard polystyrene.

Further, it is possible to previously blend a block copolymer having at least two mutually incompatible blocks (a) and (b) and comprising a conjugated diene and/or aromatic vinyl monomer with a polymer (α) compatible with the block (a) and the polymer phase (A) and/or a polymer (β) compatible with the block (b) and the polymer phase (B), which satisfies the following equations (III) and (IV):

$$S_\alpha = Mw(\alpha)/Mw(a) \leq 1.2 \qquad (III)$$

$$S_\beta = Mw(\beta)/Mw(b) \leq 1.2 \qquad (IV)$$

wherein Mw(α): weight average molecular weight of polymer (α),

Mw(β): weight average molecular weight of polymer (β),

Mw(a): weight average molecular weight of block (a) of block copolymer, and

Mw(b): weight average molecular weight of block (b) of block copolymer,
so as to improve the dispersion of the block copolymer during mixing of the rubber and obtain the better mechanical strength.

Note that S_α and S_β are preferably 0.1 to 1.2, more preferably 0.3 to 1.0.

In particular, (α) should be added when the low molecular weight portion of the polymer forming the polymer phase (A) is small and (β) should be added when the low molecular weight portion of the polymer forming the polymer phase (B) is small. Therefore, even when the above equations (I) and/or (II) are not satisfied, it is possible to obtain the effect of the present invention by blending in the polymer (α) or (β).

Here, as the polymer (α) or (β), rubbers such as IR, BR, SBR, SIBR having a suitable molecular weight are preferable, but it is not limited to a rubber so long as the object of improving the dispersion at the time of mixing the block polymer is achieved without impairing the vulcanized physical properties of the rubber composition finally obtained. Other polymers may also be used.

The amount of the polymer (α) or (β) blended should be 5 to 200 parts by weight, preferably 20 to 100 parts by weight, based upon 100 parts by weight of the block copolymer. If the amount is less than 5 parts by weight, the anticipated effect is not manifested, while if more than 200 parts by weight, the elasticity or mechanical strength is decreased, and therefore, there is a detrimental effect on the physical properties or the Mooney viscosity of the starting rubber is decreased and handling becomes difficult.

The process of production of the block copolymer used in the present invention is not particularly limited, but, for example, this may be produced by polymerizing isoprene, butadiene, or styrene monomers in a hydrocarbon solvent using an organoactive metal as an initiator. As the organoactive metal, for example, an anionic polymerizable organoactive metal such as an organoalkali metal compound, organoalkali earth metal compound, or organolanthanoid based rare earth metal compound may be mentioned. Among these, an organoalkali metal compound is particularly preferable.

According to the present invention, it is possible to further blend low molecular weight polymers (for example, IR, BR, SBR, or SIBR) as parts of the polymers forming the polymer phases so as to satisfy the above equations (I) and (II). The amounts of the low molecular weight polymers blended are preferably 1 to 50 parts by weight, based upon 100 parts by weight of the rubber

component, as a whole. If the amounts blended are too great, this leads to the decrease in the tensile strength etc., and therefore, this is not preferred.

The incompatible polymer blend comprising the polymer phases (A) and (B) used in the present invention is not particularly limited so long as two or more types of polymers selected from polymers containing conjugated dienes and/or aromatic vinyl monomers such as NR, IR, BR, SBR, are selected and constitute two incompatible polymer phases (A) and (B). Further, the block copolymer comprising the blocks (a) and (b) used in the present invention may be made any polymer provided with the above conditions. For example, a BR block, SBR block, IR block, SIR (i.e., styrene isoprene rubber) block, BIR (i.e., butadiene isoprene) block, SBIR (i.e., styrene butadiene isoprene) block, etc. may be suitably combined for use.

Representative examples of combinations of such incompatible polymers and block copolymers are as follows:

Table I

Matrix polymer (A)/(B)	Block copolymer ((a)/(b))
NR/SBR (wherein, amount of vinyl of Bd part is not more than about 60 mol%)	IR/SBR (amount of vinyl of Bd part not more than about 60 mol%) or SBR/SBR (amount of St about 20% by weight, amount of vinyl of Bd part about 70 mol%)
NR/BR (cis content not less than 90 mol%)	IR/SBR (amount of St about 20% by weight, amount of vinyl of Bd part about 50 mol%)

Of course, the present invention is not limited to the above examples.

The rubber composition according to the present invention may suitably use various conventional additives according to its application, for example, various reinforcing fillers generally used in the prior art such as carbon black and silica, softeners, antioxidant, wax, resin, vulcanization agent, vulcanization accelerator,

vulcanization accelerator activator, etc. Further, blowing agent, low moisture plasticizer, short fibers, etc. may be used.

In blending the rubber composition according to the present invention, it is preferable to first mix the rubber (i.e., matrix rubber and block copolymer) and the additives other than, for example, vulcanization agent and vulcanization accelerator according to an ordinary method, then blend them. Of course, even if some of these ingredients are separately mixed, the resultant mixture, needless to say, falls in the technical scope of the present invention so long as the object of the present invention is not impaired. Further, the blending may be carried out in any means used in the past.

The rubber composition of the present invention may be vulcanized by a general method. The amount of the above additives blended may be the general amounts. Further, the vulcanization conditions may be made the general conditions.

EXAMPLES

The present invention will be further illustrated with reference to Examples, but the present invention is of course by no means limited in scope by these Examples.

Standard Examples 1 to 6, Examples 1 to 12, and Comparative Examples 1 to 7

The ingredients of the formulations of Tables II to IV, Table V, and Table VI (parts by weight) (wherein the characteristics of the polymers used as the phase (A) and phase (B) are shown in Table VII, the characteristics of the block polymers are shown in Table VIII, and the characteristics of the polymers (α) and (β) are shown in Table IX) were mixed in 1.5 liter Bambury mixers for 4 minutes, then the vulcanization accelerators and sulfur were mixed with the mixtures by 8-inch test-use roll mill to obtain the rubber compositions. These rubber compositions were press vulcanized at 160°C for 20 minutes to prepare the desired test pieces which were

then subjected to various tests and measured in physical
properties. The physical properties of the vulcanates
obtained were as shown in Tables II, III and IV.

Mixing Method

5   The mixing methods used in the Examples and the
Comparative Examples were all according to the following
mixing specifications:

1)   Rotor speed: 60 rpm

2)   Temperature adjustment: 50°C

10   3)   Charging specifications:

0'... rubber ingredients (matrix rubber, block
copolymer)

1'... carbon black in half amount, zinc white,
stearic acid

15   2'30"... carbon black in half amount, antioxidant,
wax

3'30"... raising and lowering of ram (cleaning ram
portion)

4'00"... discharge

20   The "yes" in the compatibility section of Tables II,
III and IV indicates a compatible relationship, while the
"no" indicates an incompatible relationship.

Table II

	Standard Ex. 1	Comp. Ex. 1	Comp. Ex. 2	Ex. 1
phase (A) polymer NR-2	50	45	45	45
phase (B) polymer SBR	50	45	45	45
Block polymer BP-1	–	10	–	–
BP-2	–	–	10	–
BP-3	–	–	–	10
Compatibility				
Block (a) ↔ block (b)	–	No	No	No
Block (a) ↔ phase (A) polymer	–	Yes	Yes	Yes
Block (a) ↔ phase (B) polymer	–	No	No	No
Block (b) ↔ phase (A) polymer	–	No	No	No
Block (b) ↔ phase (B) polymer	–	Yes	Yes	Yes
Relation with molecular weight				
$S_A = Mw_{30}(A)/Mw(a)$	–	1.8	1.7	0.8
$S_B = Mw_{30}(B)/Mw(b)$	–	1.2	0.6	0.6
Physical properties of rubber composition				
Tensile strength (MPa)	23.2	23.5	24.0	26.3
Elongation (%)	370	378	380	418
Abrasion resistance index (index)	100	102	101	120

Table III

	Standard Ex. 2	Standard Ex. 3	Standard Ex. 4
phase (A) polymer NR-1	80	–	–
NR-2	–	80	–
NR-3	–	–	80
phase (B) polymer BR	20	20	20
Physical properties of rubber composition			
Tensile strength (MPa)	29.9	28.1	26.6
Elongation (%)	568	578	579
Abrasion resistance index (index)	100	100	100
Times to breakage in fatigue test	2195900	2320300	2342100

Table III (Continued)

	Comp. Ex. 3	Comp. Ex. 4	Ex. 2
phase (A) polymer NR-2	78	78	78
phase (B) polymer BR	19	19	19
Block polymer BP-4	3	-	-
BP-5	-	3	-
BP-6	-	-	3
Compatibility			
Block (a) ↔ block (b)	No	No	No
Block (a) ↔ phase (A) polymer	Yes	Yes	Yes
Block (a) ↔ phase (B) polymer	No	No	No
Block (b) ↔ phase (A) polymer	No	No	No
Block (b) ↔ phase (B) polymer	Yes	Yes	Yes
Relation with molecular weight			
$S_A = Mw_{30}(A)/Mw(a)$	1.7	1.7	0.9
$S_B = Mw_{30}(B)/Mw(b)$	0.7	0.3	0.3
Physical properties of rubber composition			
Tensile strength (MPa)	28.4	28.3	30.6
Elongation (%)	581	580	586
Abrasion resistance index (index)	101	102	106
Times to breakage in fatigue test	2310000	2298700	3212400

Table III (Continued)

	Comp. Ex. 5	Ex. 3
phase (A) polymer NR-1	78	-
NR-3	-	78
phase (B) polymer BR	19	19
Block polymer BP-5	-	3
BP-6	3	-
Compatibility		
Block (a) ↔ block (b)	No	No
Block (a) ↔ phase (A) polymer	Yes	Yes
Block (a) ↔ phase (B) polymer	No	No
Block (b) ↔ phase (A) polymer	No	No
Block (b) ↔ phase (B) polymer	Yes	Yes
Relation with molecular weight		
$S_A = Mw_{30}(A)/Mw(a)$	1.3	1.2
$S_B = Mw_{30}(B)/Mw(b)$	0.3	0.3
Physical properties of rubber composition		
Tensile strength (MPa)	29.9	29.1
Elongation (%)	572	599
Abrasion resistance index (index)	102	107
Times to breakage in fatigue test	2188800	3400200

<u>Table IV</u>

	Standard Ex. 5	Comp. Ex. 6	Ex. 4	Ex. 5
phase (A) polymer NR-1	60	58	55	57.85
NR-2	–	–	–	–
phase (B) polymer BR	40	39	39	39
Block polymer BP-7	–	3	3	3
Polymer (α) α-1	–	–	3	–
α-2	–	–	–	0.15
α-3	–	–	–	–
α-4	–	–	–	–
Polymer (β) β-1	–	–	–	–
<u>Compatibility</u>				
Block (a) ↔ block (b)	–	No	No	No
Block (a) ↔ phase (A) polymer	–	Yes	Yes	Yes
Block (a) ↔ phase (B) polymer	–	No	No	No
Block (b) ↔ phase (A) polymer	–	No	No	No
Block (b) ↔ phase (B) polymer	–	Yes	Yes	Yes
Polymer (α) ↔ block (a)	–	–	Yes	Yes
Polymer (α) ↔ phase (A) polymer	–	–	Yes	Yes
Polymer (β) ↔ block (b)	–	–	–	–
Polymer (β) ↔ phase (B) polymer	–	–	–	–
<u>Relation with molecular weight</u>				
$S_A = Mw_{30}(A)/Mw(a)$	–	2.2	2.2	2.2
$S_B = Mw_{30}(B)/Mw(b)$	–	0.3	0.3	0.3
$S_\alpha = Mw(\alpha)/Mw(a)$	–	–	0.7	1
$S_\beta = Mw(\beta)/Mw(b)$	–	–	–	–
Rate of polymers (α) and (β) added/wt% (to block polymer)	–	–	100	5
<u>Physical properties of rubber composition</u>				
Tensile strength (MPa)	27.9	28.3	29.2	28.7
Elongation (%)	550	560	588	575
Abrasion resistance index (index)	100	102	110	103

<u>Table IV (Continued)</u>

	Ex. 6	Ex. 7	Ex. 8	Comp. Ex. 7
phase (A) polymer NR-1	55	52	55	55
NR-2	–	–	–	–
phase (B) polymer BR	39	39	39	39
Block polymer BP-7	3	3	3	3
Polymer (α) α-1	–	–	–	–
α-2	3	6	–	–
α-3	–	–	3	–
α-4	–	–	–	3
Polymer (β) β-1	–	–	–	–
<u>Compatibility</u>				
Block (a) ↔ block (b)	No	No	No	No
Block (a) ↔ phase (A) polymer	Yes	Yes	Yes	Yes
Block (a) ↔ phase (B) polymer	No	No	No	No
Block (b) ↔ phase (A) polymer	No	No	No	No
Block (b) ↔ phase (B) polymer	Yes	Yes	Yes	Yes
Polymer (α) ↔ block (a)	Yes	Yes	Yes	Yes
Polymer (α) ↔ phase (A) polymer	Yes	Yes	Yes	Yes
Polymer (β) ↔ block (b)	–	–	–	–
Polymer (β) ↔ phase (B) polymer	–	–	–	–
<u>Relation with molecular weight</u>				
$S_A = Mw_{30}(A)/Mw(a)$	2.2	2.2	2.2	2.2
$S_B = Mw_{30}(B)/Mw(b)$	0.3	0.3	0.3	0.3
$S_\alpha = Mw(\alpha)/Mw(a)$	1	1	1.2	1.4
$S_\beta = Mw(\beta)/Mw(b)$	–	–	–	–
Rate of polymers (α) and (β) added/wt% (to block polymer)	100	200	100	100
<u>Physical properties of rubber composition</u>				
Tensile strength (MPa)	29.7	28.5	28.8	28.4
Elongation (%)	585	588	577	562
Abrasion resistance index (index)	108	103	105	102

Table IV (Continued)

	Ex. 9	Ex. 10	Standard Ex. 6	Ex. 11	Ex. 12
phase (A) polymer NR-1	58	56	–	–	–
NR-2	–	–	60	58	57.4
phase (B) polymer BR	38	38	40	39	39
Block polymer BP-6	–	–	–	3	3
Block polymer BP-7	3	3	–	–	–
Polymer (α) α-1	–	–	–	–	–
α-2	–	2	–	–	0.6
α-3	–	–	–	–	–
α-4	–	–	–	–	–
Polymer (β) β-1	1	1	–	–	–
Compatibility					
Block (a) ↔ block (b)	No	No	–	No	No
Block (a) ↔ phase (A) polymer	Yes	Yes	–	Yes	Yes
Block (a) ↔ phase (B) polymer	No	No	–	No	No
Block (b) ↔ phase (A) polymer	No	No	–	No	No
Block (b) ↔ phase (B) polymer	Yes	Yes	–	Yes	Yes
Polymer (α) ↔ block (a)	–	Yes	–	–	Yes
Polymer (α) ↔ phase (A) polymer	–	Yes	–	–	Yes
Polymer (β) ↔ block (b)	Yes	Yes	–	–	–
Polymer (β) ↔ phase (B) polymer	Yes	Yes	–	–	–
Relation with molecular weight					
$S_A = Mw_{30}(A)/Mw(a)$	2.2	2.2	–	0.9	0.9
$S_B = Mw_{30}(B)/Mw(b)$	0.3	0.3	–	0.3	0.3
$S_\alpha = Mw(\alpha)/Mw(a)$	–	1.0	–	–	0.6
$S_\beta = Mw(\beta)/Mw(b)$	0.3	0.3	–	–	–
Rate of polymers (α) and (β) added/wt% (to block polymer)	33	100	–	–	20
Physical properties of rubber composition					
Tensile strength (MPa)	28.4	30	26.8	27.7	29.8
Elongation (%)	570	590	520	566	602
Abrasion resistance index (index)	103	107	100	101	111

Table V: NR/SBR Blend Formulation (Parts by Weight)

Rubber component	100
Carbon black (N339)*1	50
Zinc white	3
Stearic acid	2
Antioxidant (6C)*2	3
Wax	2
Vulcanization accelerator (NS)*3	1
Sulfur	1.7

*1: Seast KH, made by Tokai Carbon Co.

*2: Santoflex 13, made by Flexis Co.

*3: Santocure NS, made by Flexis Co.

Table VI: NR/BR Blend Formulation (Parts by Weight)

Rubber component	100
Carbon black (N110)*1	50
Zinc white	5
Stearic acid	2
Antioxidant (6C)*2	3
Vulcanization accelerator (NS)*3	1.2
Sulfur	1

*1: Diablack I (made by Mitsubishi Chemical)

*2: Santoflex 13 (made by Flexis Co.)

*3: Santocure NS (made by Flexis Co.)

Table VII: Characteristics of Polymers Used as Phase (A) and Phase (B)

	Overall (Mw)	30% (Mw)
NR-1*1	7.57×10^5	3.9×10^5
NR-2*2	1.19×10^6	2.6×10^5
NR-3*3	4.65×10^5	1.8×10^5
SBR*4	3.72×10^5	1.9×10^5
BR*5	3.51×10^5	1.1×10^5

(Note)

*1: Masticated natural rubber RSS#3. Masticated by 8-inch roll mill at 80°C for 3 minutes.

*2: Natural rubber SMR-L

*3: Masticated natural rubber RSS#3. Masticated by 8-inch roll mill at 80°C for 15 minutes.

*4: NS 114 (SBR made by Nippon Zeon)

*5: BR 1220 (BR made by Nippon Zeon)

Table VIII: Characteristics of Block Polymer

	Block (a)		Block (b)	
	Microstructure	Mw	Microstructure	Mw
BP-1	Polyisoprene (cis/trans/vn=77/16/7)	1.48×10^5	SBR (St=18 wt%, Vn=11 mol%)	1.61×10^5
BP-2	Polyisoprene (cis/trans/vn=77/16/7)	1.52×10^5	SBR (St=18 wt%, Vn=11 mol%)	3.12×10^5
BP-3	Polyisoprene (cis/trans/vn=77/16/7)	3.10×10^5	SBR (St=18 wt%, Vn=11 mol%)	3.21×10^5
BP-4	Polyisoprene (cis/trans/vn=77/16/7)	1.51×10^5	SBR (St=19 wt%, Vn=46 mol%)	1.47×10^5
BP-5	Polyisoprene (cis/trans/vn=77/16/7)	1.49×10^5	SBR (St=19 wt%, Vn=46 mol%)	3.22×10^5
BP-6	Polyisoprene (cis/trans/vn=77/16/7)	3.01×10^5	SBR (St=19 wt%, Vn=46 mol%)	3.21×10^5
BP-7	Polyisoprene (cis/trans/vn=77/16/7)	1.80×10^5	SBR (St=19 wt%, Vn=46 mol%)	3.22×10^5

(Note) BP-1 to BP-7 were obtained by 2-stage polymerization by butyl lithium initiator in n-hexane solvent.

Table IX: Characteristics of Polymers Used as (α) and (β)

	Microstructure	Mw
α-1	Polyisoprene (cis/trans/vn=77/16/7)	1.2×10^5
α-2	Polyisoprene (cis/trans/vn=77/16/7)	1.8×10^5
α-3	Polyisoprene (cis/trans/vn=77/16/7)	2.2×10^5
α-4	Polyisoprene (cis/trans/vn=77/16/7)	2.5×10^5
β-1	SBR (St=19 wt%, Vn=46 mol%)	1.0×10^5

(Notes) α-1 to α-4 and β-1 were obtained by polymerization in organic solvents using organometallic compounds as initiators.

The physical properties evaluated in the above Examples were measured by the following methods:

Tensile strength (Mpa): Measured according to JIS K6251.

Elongation (%): Measured according to JIS K6251.

Abrasion resistance test: Measured using a Lambourn abrasion tester at conditions of a slip rate of 25% and a load of 5 kg. The results are shown indexed to the formulation of the corresponding Standard Examples as 100

(abrasion resistance index). The larger the figure, the better the abrasion resistance shown.

Times to breakage in fatigue test: Shown by number of times to breakage of a JIS No. 3 dumbbell shaped sample after being given repeated deformation at a cycle rate of 400 rpm at an elongation stress of 100% (average for four tests).

The incompatibility of polymers was judged by the following method:

1) The incompatibility of the polymer phases (A) and (B) of the polymer blend was judged by vulcanizing the polymer blend, preparing ultrathin slice samples by the freezing method, then dyeing these in a gas phase with a benzene solution of osmium tetraoxide at room temperature for about 15 hours. The presence of phase (a) separated structure was examined by observation through a transmission type electron microscope at magnifications of about 5000 to 10,000.

2) The incompatibility of the blocks (a) and (b) of the block copolymer was judged by preparing samples in the same way as above from the block copolymer in the unvulcanized state, then observing them through a transmission type electron microscope at a magnification of about 60,000 to examine the presence of a phase separated structure.

3) The incompatibility of the blocks of the block copolymer and the polymer phases of the polymer blend was judged by separating polymerizing and preparing the polymers corresponding to the polymers constituting the blocks, mixing these with the matrix polymers, vulcanizing them, then proceeding in the same way as above to prepare samples for observation through an electron microscope and observing these at magnifications of about 5000 to 10,000 to examine the presence of phase separated structures.

In addition, the compatibilities and incompatibilities may be decided by judging the presence

of bimodal or not from the temperature dependence curve
of tan δ or by judging the presence of plurality of glass
transition temperatures or not of the blend polymers can
be observed by DSC measurement and further may be judged

5   by an optical microscope if the phase separated structure
reaches as much as several dozen microns. Among these,
the above direct observation by an electron microscope is
the most sensitive method.

As explained above and as shown in Examples 1 to 12,

10   the rubber compositions according to the present
invention are improved in mechanical strength such as
tensile strength, elongation, abrasion resistance, and
fatigue resistance compared with the rubber compositions
of Comparative Examples 1 to 7.

CLAIMS

1.   A rubber composition comprising (i) an incompatible polymer blend comprising at least two diene rubbers selected from the group consisting of rubbers containing at least one conjugated diene monomer and, optionally, at least one aromatic vinyl monomer and forming two imcompatible polymer phases (A) and (B) and (ii) 0.1 to 20 parts by weight, based upon 100 parts by weight of the total polymer component including the block copolymer, of a block copolymer having at least two mutually incompatible blocks (a) and (b) in which the block (a) is compatible with the polymer phase (A) and incompatible with the polymer phase (B) and the block (b) is compatible with the polymer phase (B) and incompatible with the polymer phase (A), and comprising at least one conjugated diene monomer and, optionally, at least one aromatic vinyl monomer, wherein the molecular weights of the polymers forming the polymer phases (A) and (B) satisfy the following equations (I) and (II):

$$S_A = Mw_{30}(A)/Mw(a) \leq 1.2 \qquad (I)$$

$$S_B = Mw_{30}(B)/Mw(b) \leq 1.2 \qquad (II)$$

wherein $Mw_{30}(A)$: molecular weight of the low molecular weight portion of the polymer forming the polymer phase (A),

$Mw_{30}(B)$: molecular weight of the low molecular weight portion of the polymer forming the polymer phase (B),

$Mw(a)$: weight average molecular weight of block (a) of block copolymer, and

$Mw(b)$: weight average molecular weight of block (b) of block copolymer.

2.   A rubber composition as claimed in claim 1, wherein 5 to 200 parts by weight, based upon 100 parts by weight of the block copolymer, of polymer (α) compatible with the block (a) and the polymer phase (A) and/or polymer (β) compatible with the block (b) and polymer

phase (B) are further blended and the weight average
molecular weights of the polymers (α) and (β) satisfy the
following equations (III) and (IV):

$$S_\alpha = Mw(\alpha)/Mw(a) \leq 1.2 \qquad\qquad\qquad (III)$$

$$S_\beta = Mw(\beta)/Mw(b) \leq 1.2 \qquad\qquad\qquad (IV)$$

wherein Mw(α): weight average molecular weight of
polymer (α),

Mw(β): weight average molecular weight of polymer
(β),

Mw(a): weight average molecular weight of block (a)
of block copolymer, and

Mw(b): weight average molecular weight of block (b)
of block copolymer.

   3.   A rubber composition as claimed in claim 1,
wherein said diene rubbers are NR, IR, BR, SBR, SIR and
SIBR.

   4.   A rubber composition as claimed in claim 3,
wherein a weight ratio of polymer phase (A)/polymer phase
(B) is 90/10 to 10/90.

   5.   A rubber composition as claimed in claim 1,
wherein said block copolymer contains at least two blocks
selected from the group consisting of BR block, SBR
block, IR block, SIR block, BIR block and SBIR block.

   6.   A rubber composition as claimed in claim 5,
wherein a weight ratio of block (a)/block (b) is 80/20 to
20/80.

   7.   A rubber composition as claimed in claim 2,
wherein said polymers (α) and (β) are selected from IR,
BR, SBR and SIBR.

   8.   A rubber composition comprising (i) an
incompatible polymer blend comprising at least two diene-
based rubbers selected from the group consisting of
rubbers containing at least one conjugated diene monomer
and, optionally, at least one aromatic vinyl monomer and
forming two polymer phases (A) and (B) and (ii) 0.1 to 20

parts by weight, based upon 100 parts by weight of the
total polymer component including the block copolymer, of
block copolymer having at least two mutually incompatible
blocks (a) and (b), in which the block (a) is compatible
5   with the polymer phase (A) and incompatible with the
polymer phase (B) and the block (b) is compatible with
the polymer phase (B) and incompatible with the polymer
phase (A), and comprising at least one conjugated diene
monomer and, optionally, at least one aromatic vinyl
10 monomer and (iii) 5 to 200 parts by weight, based upon
100 parts by weight of the block copolymer, of at least
one polymer selected from the group consisting of a
polymer (α) compatible with the block (a) and the
polymer phase (A) and a polymer (β) compatible with the
15 block (b) and polymer phase (B), wherein the weight
average molecular weights of the polymers (α) and (β)
satisfy the following equations (III) and (IV):

$$S_\alpha = Mw(\alpha)/Mw(a) \leq 1.2 \qquad\qquad (III)$$

$$S_\beta = Mw(\beta)/Mw(b) \leq 1.2 \qquad\qquad (IV)$$

20 wherein $Mw(\alpha)$: weight average molecular weight of
polymer (α),

$Mw(\beta)$: weight average molecular weight of polymer
(β),

$Mw(a)$: weight average molecular weight of block (a)
25 of block copolymer, and

$Mw(b)$: weight average molecular weight of block (b)
of block copolymer.

9.   A rubber composition as claimed in claim 8,
wherein said diene rubbers are NR, IR, BR, SBR, SIR and
30 SIBR.

10. A rubber composition as claimed in claim 9,
wherein a weight ratio of polymer phase (A)/polymer phase
(B) is 90/10 to 10/90.

11. A rubber composition as claimed in claim 8,

wherein said block copolymer contains at least two blocks
selected from the group consisting of BR block, SBR
block, IR block, SIR block, BIR block and SBIR block.

12. A rubber composition as claimed in claim 11,
wherein a weight ratio of block (a)/block (b) is 80/20 to
20/80.

13. A rubber composition as claimed in claim 8,
wherein said polymers (α) and (β) are selected from IR,
BR, SBR and SIBR.

14. A rubber composition comprising 100 parts by
weight of a block copolymer having at least two mutually
incompatible blocks (a) and (b) and composed of at least
one conjugated diene monomer and, optionally, at least
one aromatic vinyl monomer and 5 to 200 parts by weight
of at least one polymer selected from the group
consisting of a polymer (α) compatible with the block
(a) and a polymer (β) compatible with the block (b),
wherein the weight average molecular weights of the
polymers (α) and (β) satisfy the following equations
(III) and (IV):

$$S_\alpha = Mw(\alpha)/Mw(a) \leq 1.2 \qquad (III)$$

$$S_\beta = Mw(\beta)/Mw(b) \leq 1.2 \qquad (IV)$$

wherein $Mw(\alpha)$: weight average molecular weight of
polymer (α),

$Mw(\beta)$: weight average molecular weight of polymer
(β),

$Mw(a)$: weight average molecular weight of block (a)
of block copolymer, and

$Mw(b)$: weight average molecular weight of block (b)
of block copolymer.

15. A rubber composition as claimed in claim 14,
wherein said block copolymer contains at least two blocks
selected from the group consisting of BR block, SBR
block, IR block, SIR block, BIR block and SBIR block.

16.   A rubber composition as claimed in claim 15, wherein a weight ratio of block (a)/block (b) is 80/20 to 20/80.

17.   A rubber composition as claimed in claim 14, 5   wherein said polymers (α) and (β) are selected from IR, BR, SBR and SIBR.

RUBBER COMPOSITION

5

ABSTRACT OF THE DISCLOSURE

A rubber composition comprising (i) an incompatible
polymer blend of at least two diene rubbers selected from
10 rubbers containing a conjugated diene and, optionally, an
aromatic vinyl monomer and forming two polymer phases (A)
and (B), and (ii) 0.1 to 20 parts by weight, based upon
100 parts by weight of the total polymer component
including the block copolymer, of a block copolymer
15 having at least two mutually incompatible blocks (a) and
(b), wherein the block (a) is compatible with the polymer
phase (A) and incompatible with the polymer phase (B) and
the block (b) is compatible with the polymer phase (B)
and incompatible with the polymer phase (A), and composed
20 of a conjugated diene and, optionally, an aromatic vinyl
monomer, and wherein the molecular weights of the
polymers forming the polymer phases (A) and (B) satisfy
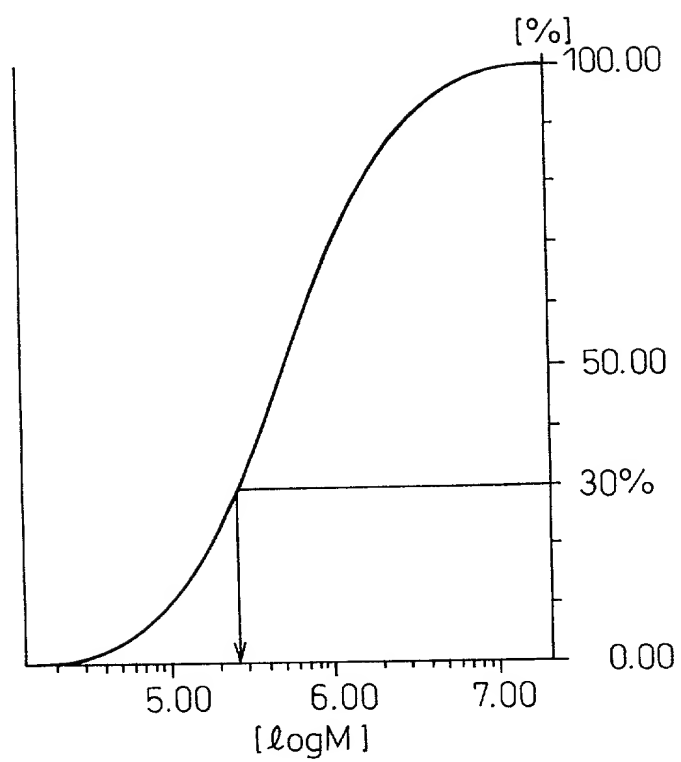the specified equations (I) and (II) mentioned in the
specification.
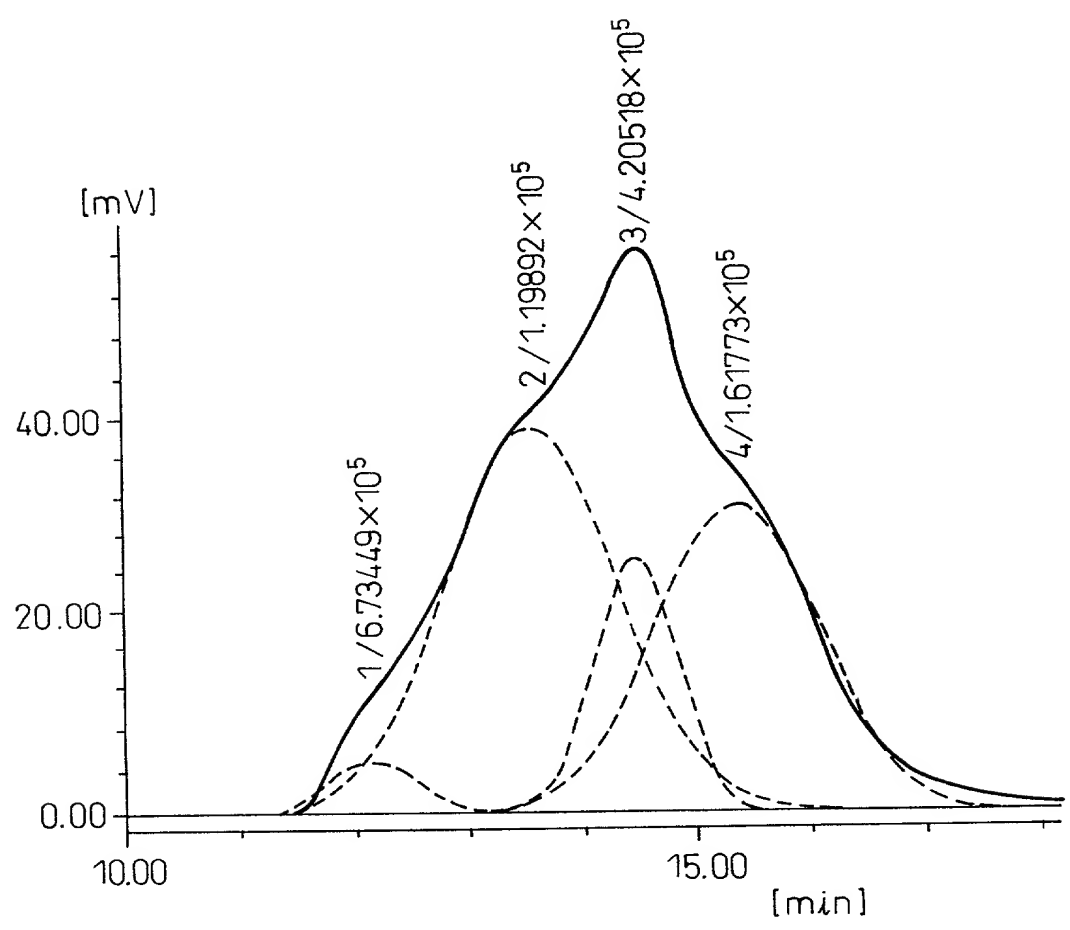
Fig.1

Fig. 2

Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

YR, NZ-G432
-US

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

RUBBER COMPOSITION

_____

_____

上記発明の明細書（下記の欄でx印がついていない場合は、本書に添付）は、

the specification of which is attached hereto unless the following box is checked:

☐ __月__日に提出され、米国出願番号または特許協定条約国際出願番号を_____とし、（該当する場合）_____ に訂正されました。

☐ was filed on _____
as United States Application Number or
PCT International Application Number
_____ and was amended on
_____ (if applicable).

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

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I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

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I hereby claim foreign priority under Title 35, United States Code, Section 119 (a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)
外国での先行出願

			Priority Not Claimed 優先権主張なし
11-050710 (Pat. Appln.) (Number) (番号)	Japan (Country) (国名)	26/February/1999 (Day/Month/Year Filed) (出願年月日)	二
2000-41396 (Pat. Appln.) (Number) (番号)	Japan (Country) (国名)	15/February/2000 (Day/Month/Year Filed) (出願年月日)	二

私は、第３５編米国法典１１９条 (e) 項に基いて下記の米国特許出願規定に記載された権利をここに主張いたします。

I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below.

(Application No.) (出願番号)	(Filing Date) (出願日)	(Application No.) (出願番号)	(Filing Date) (出願日)

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(Application No.) (出願番号)	(Filing Date) (出願日)	(Status: Patented, Pending, Abandoned) (現況：特許許可済、係属中、放棄済)
(Application No.) (出願番号)	(Filing Date) (出願日)	(Status: Patented, Pending, Abandoned) (現況：特許許可済、係属中、放棄済)

私は、私自身の知識に基ずいて本宣言書中で私が行なう表明が真実であり、かつ私の入手した情報と私の信じるところに基ずく表明が全て真実であると信じていること、さらに故意になされた虚偽の表明及びそれと同等の行為は米国法典第１８編第１００１条に基ずき、罰金または拘禁、もしくはその両方により処罰されること、そしてそのような故意による虚偽の声明を行なえば、出願した、又は既に許可された特許の有効性が失われることを認識し、よってここに上記のごとく宣誓を致します。

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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POWER OF ATTORNEY: As a named inventor. I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith *(list name and registration number)*

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第二共同発明者　　　日付	Second inventor's signature　　　Date *Masayuki Kawazoe*　February 10, 2000
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（第三以降の共同発明者についても同様に記載し、署名をすること）

(Supply similar information and signature for third and subsequent joint inventors.)

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第四共同発明者名		Full name of fourth joint inventor, if any	
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国籍		Citizenship	
私書箱		Post Office Address	
第五共同発明者名		Full name of fifth joint inventor, if any	
第五共同発明者の署名	日付	Fifth inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	
第六共同発明者名		Full name of sixth joint inventor, if any	
第六共同発明者の署名	日付	Sixth inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	